

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant	:	Shoji, et al.
Appl. No.	:	10/642,952
Filed	:	August 18, 2003
For	:	PROCESS FOR PRODUCING DIMETHYL ETHER
Examiner	:	Rosalynd Ann Keys
Group Art Unit	:	4157

DECLARATION UNDER RULE 132

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Dear Sir:

I, Satoshi Terai, do hereby declare as follows:

1. I am a co-inventor of the above-identified application.
2. The following experiments were conducted by me or under my supervision or control to evaluate a reduction in conversion rate of methanol to dimethyl ether (DME) after six months of operation using activated alumina catalysts.
3. Five (5) mL of a  $\gamma$ -alumina catalyst having a specific surface area (SA), a pore volume (PV), an average pore radius (R), and a  $\text{Na}_2\text{O}$  content shown in a table below (measured by the methods described on page 14 of the specification) were loaded in a stainless fixed bed reactor with an inner diameter of 16 mm equipped with an electric furnace around its periphery (no additional active component was added to the catalyst). The  $\text{Na}_2\text{O}$  content of the  $\gamma$ -alumina catalysts were reduced by either selecting low-Na content raw materials or conducting acid washing. At a reaction temperature of 270°C, methanol was then fed at a GHSV of 1700 h<sup>-1</sup> and a pressure of 0 MPa-G (gauge pressure), while methanol was heated to its evaporating temperature at the reaction pressure in an upstream line of the reactor so that methanol was vaporized before entering the catalyst layer. During the process, a temperature of the catalyst layer was 270°C. Under these conditions, a conversion ratio of methanol to DME after one year

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of operation was measured for one of catalysts No. 1 to No. 4 shown in the table below. A relative activity of each catalyst was calculated with reference to catalyst No. 1 by dividing the conversion ratio of each catalyst by that of catalyst No. 1. The results are shown in the table below.

4. Table

Catalyst No.	R [nm]	PV [ml/g]	SA [m <sup>2</sup> /g]	Na <sub>2</sub> O [wt%]	Relative Activity after one year [-]
1	3.9	0.404	208	0.002	1.00
2	4.0	0.363	183	0.033	1.03
3	5.5	0.400	145	0.030	0.41
4	6.6	0.760	229	0.038	0.27

5. As can be seen from the table, the average pore radius (R) predominantly controlled long term stability of activity, as compared with the other characteristics, and when the average pore radius was 5.5 nm or 6.6 nm, long term stability of activity was drastically dropped.

6. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statement may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

Dated: January 11, 2007

By: Satoshi Terai  
Satoshi Terai